IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

I, ADRIAN PAUL BROWN, M.A., M.I.L., M.I.T.I., declare

1. That I am a citizen of the United Kingdom of Great Britain and Northern Ireland, residing at 5 Gilbert Road, London, SE11 4NZ.

2. That I am well acquainted with the German and English languages.

3. That the attached is a true translation into the English language of the certified copy of Swiss Patent Application No. 2002 1956/02 as filed on 21st November 2002.

4. That all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of the patent application in the United States of America or any patent issuing thereon.

DECLARED THIS 11th DAY OF APRIL 2005

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Herbicidal composition

The present invention relates to a novel herbicidal synergistic composition comprising a herbicidal active ingredient combination that is suitable for the selective control of weeds in crops of useful plants, for example in crops of rice.

The invention relates also to a method of controlling weeds in crops of useful plants and to the use of the novel composition for that purpose.

The compounds mesotrione (500), sulcotrione (664), isoxaflutole (436), pyrazoxyfen (666), pyrazolynate (663), benzofenap (71), sulfentrazone (711), pyraflufen-ethyl (662), beflubutamid (57), cafenstrole (108), dimethametryn (253), clomeprop (160), prometryn (641), cinosulfuron (154), triasulfuron (773), prosulfuron (657), imazosulfuron (444), ethoxysulfuron (307), sulfosulfuron (714), iodosulfuron (454), benzobicyclon (70), bentazone (69), simetryn (699), bensulfuron (66), pyrazosulfuron (665), metsulfuron (536), azimsulfuron (45), clodinafop (156), 2,4-D (205), acetochlor (7), metolachlor (529), S-metolachlor (530), and agronomically acceptable salts thereof, exhibit herbicidal action, as is described, for example, in The Pesticide Manual, 12th Edition (BCPC), 2000.
Tritosulfuron, registered as no. 142469-14-5 in CAS (Chemical Abstracts), is known from EP-A-559 814 and WO 01/24633. The herbicidal action of that compound is also described therein.
The herbicidal action of metamifop is known, for example, from WO 00/05956.
Trifloxysulfuron and its herbicidal action are described, for example, in WO 00/52006.
The compound 2-[[[[(4,6-dimethoxy-2-pyrimidinyl)amino]carbonyl]amino]sulfonyl]-4-[[(methylsulfonyl)amino]methyl]benzoic acid is known under the name mesosulfuron. Its herbicidal action is described in EP-A-559 814 and WO 01/24633.
The compound N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide and its herbicidal action are known from WO 02/30921.
Pyraclonil (1-(3-chloro-4,5,6,7-tetrahydropyrazolo[1,5-*a*]pyridin-2-yl)-5-(methyl-2-propynylamino)-1*H*-pyrazole-4-carbonitrile, registered as RN 158353-15-2 in CAS (Chemical Abstracts)), is known from WO 94/08999. The herbicidal action of that compound is also described therein.

Surprisingly, it has now been found that a combination of variable amounts of metamifop with at least one active ingredient from the above listing exhibits a synergistic action that is capable of controlling, both pre-emergence and post-emergence, the majority of weeds occurring especially in crops of useful plants, without appreciably damaging the useful plants.

There is therefore proposed in accordance with the present invention a novel synergistic composition for the selective control of weeds which comprises a mixture of
a) metamifop and
b) a synergistically effective amount of at least one compound selected from the compounds of the group mesotrione, sulcotrione, isoxaflutole, pyrazoxyfen, pyrazolynate, benzofenap, sulfentrazone, pyraflufen-ethyl, beflubutamid, cafenstrole, dimethametryn, clomeprop, prometryn, cinosulfuron, triasulfuron, prosulfuron, imazosulfuron, ethoxysulfuron, sulfosulfuron, iodosulfuron, tritosulfuron, mesosulfuron, trifloxysulfuron, benzobicyclon, acetochlor, metolachlor, S-metolachlor, pyraclonil and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, (bentazone and trifloxysulfuron), (bentazone and ethoxysulfuron), (bentazone and mesosulfuron), (bentazone and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide), (simetryn and cinosulfuron), (simetryn and triasulfuron), (simetryn and prosulfuron), (simetryn and trifloxysulfuron), (simetryn and imazosulfuron), (simetryn and ethoxysulfuron), (simetryn and sulfosulfuron), (simetryn and iodosulfuron), (simetryn and mesosulfuron), (simetryn and tritosulfuron) and (simetryn and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide) and (clodinafop and 2,4-D), the two-component mixture of metamifop with benzobicyclon being excluded.

It is extremely surprising that combinations of those active ingredients exceed the additive effect on the weeds to be controlled that is to be expected in principle and thus broaden the range of action of both active ingredients especially in two respects: firstly, the rates of application of the individual compounds are reduced while a good level of action is maintained and, secondly, the composition according to the invention achieves a high level of weed control also in those cases where the individual substances, in the low rates of application range, have become useless from the agronomic standpoint. The result is a considerable broadening of the spectrum of weeds and an additional increase in selectivity in respect of the crops of useful plants, as is necessary and desirable in the event of an

unintentional overdose of active ingredient. The composition according to the invention, while retaining excellent control of weeds in crops of useful plants, also allows greater flexibility in succeeding crops.

The composition according to the invention can be used against a large number of agronomically important weeds, such as Stellaria, Nasturtium, Agrostis, Digitaria, Avena, Setaria, Sinapis, Lolium, Solanum, Bromus, Apera, Alopecurus, Matricaria, Abutilon, Sida, Xanthium, Amaranthus, Chenopodium, Ipomoea, Chrysanthemum, Galium, Viola and Veronica. The composition according to the invention is suitable for all methods of application conventionally used in agriculture, e.g. pre-emergence application, post-emergence application and seed dressing. The composition according to the invention is suitable for controlling weeds in rice. "Crops of useful plants" are to be understood to mean also those which have been made tolerant to herbicides or classes of herbicides as a result of conventional methods of breeding or genetic engineering methods.

The composition according to the invention comprises the mentioned active ingredients in any mixing ratio, but usually has an excess of one component over the other. Preferred mixing ratios of the active ingredients are from 100:1 to 1:100 and 50:1 to 1:50.

Compositions that have been found to be especially effective are the combinations metamifop and sulcotrione, metamifop and isoxaflutole, metamifop and pyrazoxyfen, metamifop and pyrazolynate, metamifop and benzofenap, metamifop and sulfentrazone, metamifop and pyraflufen-ethyl, metamifop and beflubutamid, metamifop and cafenstrole, metamifop and dimethametryn, metamifop and clomeprop, metamifop and prometryn, metamifop and trifloxysulfuron, metamifop and mesotrione and cinosulfuron, metamifop and sulcotrione and cinosulfuron, metamifop and isoxaflutole and cinosulfuron, metamifop and pyrazoxyfen and cinosulfuron, metamifop and pyrazolynate and cinosulfuron, metamifop and benzofenap and cinosulfuron, metamifop and sulfentrazone and cinosulfuron, metamifop and pyraflufen-ethyl and cinosulfuron, metamifop and beflubutamid and cinosulfuron, metamifop and cafenstrole and cinosulfuron, metamifop and dimethametryn and cinosulfuron, metamifop and clomeprop and cinosulfuron, metamifop and prometryn and cinosulfuron, metamifop and trifloxysulfuron and cinosulfuron, metamifop and benzobicyclon and cinosulfuron, metamifop and mesotrione and triasulfuron, metamifop and sulcotrione and triasulfuron, metamifop and isoxaflutole and triasulfuron, metamifop and pyrazoxyfen and triasulfuron, metamifop and pyrazolynate and triasulfuron, metamifop and benzofenap and triasulfuron, metamifop and sulfentrazone and triasulfuron, metamifop and pyraflufen-

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and isoxaflutole and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and pyrazoxyfen and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and pyrazolynate and N-[(4,6-dimethoxypyrimidin-2-yl)amino-carbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and benzofenap and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and sulfentrazone and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and pyraflufen-ethyl and N-[(4,6-dimethoxypyrimidin-2-yl)amino-carbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and beflubutamid and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and cafenstrole and N-[(4,6-dimethoxy-pyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfon-amide, metamifop and dimethametryn and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and clomeprop and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and prometryn and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and trifloxysulfuron and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and benzobicyclon and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and bentazone and trifloxysulfuron, metamifop and bentazone and ethoxysulfuron, metamifop and bentazone and mesosulfuron, metamifop and bentazone and N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfonamide, metamifop and simetryn and cinosulfuron, metamifop and simetryn and triasulfuron, metamifop and simetryn and prosulfuron, metamifop and simetryn and trifloxysulfuron, metamifop and simetryn and imazosulfuron, metamifop and simetryn and ethoxysulfuron, metamifop and simetryn and sulfosulfuron, metamifop and simetryn and iodosulfuron, metamifop and simetryn and mesosulfuron, metamifop and simetryn and tritosulfuron, metamifop and simetryn and N-[(4,6-dimethoxy-pyrimidin-2-yl)aminocarbonyl]-2-(2-fluoro-1-methoxy-acetoxy-n-propyl)pyridine-3-sulfon-amide, and also metamifop and clodinafop and 2,4-D.

Compositions that have been found to be very especially effective are the combinations metamifop and acetochlor, metamifop and metolachlor, metamifop and S-metolachlor, metamifop and pyraclonil, metamifop and mesotrione.

The rate of application may vary within wide limits and depends on the nature of the soil, the method of application (pre- or post-emergence; seed dressing; application to the seed furrow; no tillage application etc.), the crop plant, the weed to be controlled, the prevailing climatic conditions, and other factors governed by the method of application, the time of application and the target crop. The active ingredient mixture according to the invention can generally be applied at a rate of from 0.001 to 1.5 kg of active ingredient mixture per hectare.

The mixtures according to the invention may be used in unmodified form, that is to say as obtained in synthesis. Preferably, however, they are formulated in customary manner, together with the adjuvants conventionally used in formulation technology, such as solvents, solid carriers or surfactants, for example into emulsifiable concentrates, directly sprayable or dilutable solutions, wettable powders, soluble powders, dusts, granules or microcapsules, as is described in WO 97/34483, pages 9 to 13. As with the nature of the compositions, the methods of application, such as spraying, atomising, dusting, wetting, scattering or pouring, are chosen in accordance with the intended objectives and the prevailing circumstances. The formulations, i.e. the compositions, preparations or products comprising the mixtures according to the invention, and also, where appropriate, one or more solid or liquid formulation adjuvants, are prepared in a manner known *per se*, e.g. by intimately mixing and/or grinding the active ingredients with the formulation adjuvants, e.g. solvents or solid carriers. In addition, surface-active compounds (surfactants) may also be used in the preparation of the formulations.

Examples of solvents and solid carriers are given, for example, in WO 97/34485, page 6. Depending on the nature of the active ingredients to be formulated, suitable surface-active compounds are non-ionic, cationic and/or anionic surfactants and surfactant mixtures having good emulsifying, dispersing and wetting properties. Examples of suitable anionic, non-ionic and cationic surfactants are listed, for example, in WO 97/34485, pages 7 and 8. Also suitable for the preparation of the herbicidal compositions according to the invention are the surfactants conventionally used in formulation technology, which are described, *inter alia*, in "McCutcheon's Detergents and Emulsifiers Annual" MC Publishing Corp., Ridgewood New Jersey, 1981, Stache, H., "Tensid-Taschenbuch", Carl Hanser Verlag, Munich/Vienna, 1981 and M. and J. Ash, "Encyclopedia of Surfactants", Vol I-III, Chemical Publishing Co., New York, 1980-81.

The herbicidal formulations usually contain from 0.1 to 99 % by weight, especially from 0.1 to 95 % by weight, of active ingredient mixture, from 1 to 99.9 % by weight of a solid or liquid formulation adjuvant, and from 0 to 25 % by weight, especially from 0.1 to 25 % by weight, of a surfactant.

Whereas the preferred commercial products are usually concentrates, the end user will normally employ dilute formulations. The compositions may also comprise further ingredients, such as stabilisers, e.g. vegetable oils or epoxidised vegetable oils (epoxidised coconut oil, rapeseed oil or soybean oil), antifoams, e.g. silicone oil, preservatives, viscosity regulators, binders, tackifiers, and also fertilisers or other active ingredients. Preferred formulations have especially the following compositions:
(% = percent by weight)

Emulsifiable concentrates:

active ingredient mixture:	from 1 to 90 %, preferably from 5 to 20 %
surface-active agent:	from 1 to 30 %, preferably from 10 to 20 %
liquid carrier:	from 5 to 94 %, preferably from 70 to 85 %

Dusts:

active ingredient mixture:	from 0.1 to 10 %, preferably from 0.1 to 5 %
solid carrier:	from 99.9 to 90 %, preferably from 99.9 to 99 %

Suspension concentrates:

active ingredient mixture:	from 5 to 75 %, preferably from 10 to 50 %
water:	from 94 to 24 %, preferably from 88 to 30 %
surface-active agent:	from 1 to 40 %, preferably from 2 to 30 %

Wettable powders:

active ingredient mixture:	from 0.5 to 90 %, preferably from 1 to 80 %
surface-active agent:	from 0.5 to 20 %, preferably from 1 to 15 %
solid carrier:	from 5 to 95 %, preferably from 15 to 90 %

Granules:

active ingredient mixture:	from 0.1 to 30 %, preferably from 0.1 to 15 %
solid carrier:	from 99.5 to 70 %, preferably from 97 to 85 %

The Examples that follow illustrate the invention further. They do not limit the invention.

F1. Emulsifiable concentrates	a)	b)	c)	d)
active ingredient mixture	5 %	10 %	25 %	50 %
calcium dodecylbenzenesulfonate	6 %	8 %	6 %	8 %
castor oil polyglycol ether (36 mol of ethylene oxide)	4 %	-	4 %	4 %
octylphenol polyglycol ether (7-8 mol of ethylene oxide)	-	4 %	-	2 %
cyclohexanone	-	-	10 %	20 %
aromatic C_9-C_{12}hydrocarbon mixture	85 %	78 %	55 %	16 %

Emulsions of any desired concentration can be prepared from such concentrates by dilution with water.

F2. Solutions	a)	b)	c)	d)
active ingredient mixture	5 %	10 %	50 %	90 %
1-methoxy-3-(3-methoxy-propoxy)-propane	-	20 %	20 %	-
polyethylene glycol (mol. wt. 400)	20 %	10 %	-	-
N-methyl-2-pyrrolidone	-	-	30 %	10 %
aromatic C_9-C_{12}hydrocarbon mixture	75 %	60 %	-	-

The solutions are suitable for application in the form of micro-drops.

F3. Wettable powders	a)	b)	c)	d)
active ingredient mixture	5 %	25 %	50 %	80 %
sodium lignosulfonate	4 %	-	3 %	-
sodium lauryl sulfate	2 %	3 %	-	4 %
sodium diisobutylnaphthalenesulfonate	-	6 %	5 %	6 %
octylphenol polyglycol ether (7-8 mol of ethylene oxide)	-	1 %	2 %	-
highly dispersed silicic acid	1 %	3 %	5 %	10 %
kaolin	88 %	62 %	35 %	-

The active ingredient is mixed thoroughly with the adjuvants and the mixture is thoroughly ground in a suitable mill, affording wettable powders which can be diluted with water to give suspensions of any desired concentration.

F4. Coated granules	a)	b)	c)
active ingredient mixture	0.1 %	5 %	15 %
highly dispersed silicic acid	0.9 %	2 %	2 %
inorganic carrier material (diameter 0.1 - 1 mm) for example $CaCO_3$ or SiO_2	99.0 %	93 %	83 %

The active ingredient is dissolved in methylene chloride, the solution is sprayed onto the carrier, and the solvent is subsequently evaporated off *in vacuo*.

F5. Coated granules	a)	b)	c)
active ingredient mixture	0.1 %	5 %	15 %
polyethylene glycol (mol. wt. 200)	1.0 %	2 %	3 %
highly dispersed silicic acid	0.9 %	1 %	2 %
inorganic carrier material (diameter 0.1 - 1 mm) for example $CaCO_3$ or SiO_2	98.0 %	92 %	80 %

The finely ground active ingredient is uniformly applied, in a mixer, to the carrier material moistened with polyethylene glycol, yielding non-dusty coated granules.

F6. Extruder granules	a)	b)	c)	d)
active ingredient mixture	0.1 %	3 %	5 %	15 %
sodium lignosulfonate	1.5 %	2 %	3 %	4 %
carboxymethylcellulose	1.4 %	2 %	2 %	2 %
kaolin	97.0 %	93 %	90 %	79 %

The active ingredient is mixed with the adjuvants, and the mixture is ground, moistened with water, extruded and then dried in a stream of air.

F7. Dusts	a)	b)	c)
active ingredient mixture	0.1 %	1 %	5 %
talcum	39.9 %	49 %	35 %
kaolin	60.0 %	50 %	60 %

Ready-to-use dusts are obtained by mixing the active ingredient with the carriers and grinding the mixture in a suitable mill.

F8. Suspension concentrates	a)	b)	c)	d)
active ingredient mixture	3 %	10 %	25 %	50 %
ethylene glycol	5 %	5 %	5 %	5 %
nonylphenol polyglycol ether (15 mol of ethylene oxide)	-	1 %	2 %	-
sodium lignosulfonate	3 %	3 %	4 %	5 %
carboxymethylcellulose	1 %	1 %	1 %	1 %
37 % aqueous formaldehyde solution	0.2 %	0.2 %	0.2 %	0.2 %
silicone oil emulsion	0.8 %	0.8 %	0.8 %	0.8 %
water	87 %	79 %	62 %	38 %

The finely ground active ingredient is intimately mixed with the adjuvants, giving a suspension concentrate from which suspensions of any desired concentration can be obtained by dilution with water.

It is often more practical to formulate the active ingredients of the mixtures according to the invention separately and then, shortly before application, to bring them together in the applicator in the desired mixing ratio in the form of a "tank mixture" in water.

Biological Examples:

Example B1: Pre-emergence test:

The test plants are sown in pots under greenhouse conditions. A standard soil is used as cultivation substrate. At a pre-emergence stage, the herbicides, both on their own and in admixture, are applied to the surface of the soil. The rates of application depend on the optimum concentrations ascertained under field conditions or greenhouse conditions. The tests are evaluated after from 2 to 4 weeks (100 % action = plant is completely dead; 0 % action = no phytotoxic action). The mixtures used in this test show good results.

Example B2: Post-emergence test:

The test plants are cultivated in pots under greenhouse conditions until a post-application stage. A standard soil is used as cultivation substrate. At a post-emergence stage, the herbicides, both on their own and in admixture, are applied to the test plants. The rates of application depend on the optimum concentrations ascertained under field conditions or greenhouse conditions. The tests are evaluated after from 2 to 4 weeks (100 % action = plant is completely dead; 0 % action = no phytotoxic action). The mixtures used in this test show good results.